

Interactive comment on “Estimates of mercury flux into the United States from non-local and global sources: results from a 3-D CTM simulation” by B. A. Drewniak et al.

Anonymous Referee #2

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General Comments

This paper addresses an issue of importance in the global mercury budget, namely the implications of different estimates of Chinese sources on U.S. mercury concentration and deposition. The authors apply a well-known global chemical transport model (MOZART) which previously had not been used for mercury chemistry. This in itself is an advance, as only a few mercury models exist and have been compared (see Bullock et al., 2008).

However, the paper has two fundamental problems in its analysis and application of mercury models to this task. First, it lacks sufficient elaboration and benchmarking of

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model results against measured data, which are available both in the U.S. and China. The comparisons that are included do not show enough primary data to establish confidence in the model simulation, especially for a first application of a new mercury simulation. Second, the paper does not put its result into sufficient context given the multiple other studies that have dealt with mercury emissions from Asia and long-range transport of Asian emissions more generally. Thus, I am not convinced that this paper presents any new or interesting results beyond the application of a different mercury model than used before (where two others have done similar calculations). A related problem is that the paper does not address the uncertainty in Asian land sources of Hg(0). With a simple chemical mechanism and little discussion of fundamental processes, it is difficult to see how the result would be different in the case of a simple inert tracer with appropriate lifetime. These comments are elaborated further under specific comments below.

Specific Comments (by paper section)

1. Introduction: the authors could use more discussion of previous studies that quantify Asian long-range transport (of mercury and other substances). The authors note the study of Seigneur et al. that Asian anthropogenic emission of Hg contributed 21% of total Hg deposition in the contiguous US in 1998. In addition the authors should discuss Strode et al. (2008), who found that Asian anthropogenic sources contributed 15% in the Western U.S. and 12% in the eastern U.S. Some discussion of other long-range transport studies of mercury and other species would be warranted here.

2.2 Mercury Chemistry: the authors should at least acknowledge Calvert and Lindberg (2005) and the likelihood that the reactions with OH and O₃ do not occur in the atmosphere, and discuss how using e.g. Br as an oxidant may change their results if at all. (I suspect not much, but it should be addressed). They should additionally acknowledge and discuss how omitting Hg(0) dry deposition may change results (see Lin et al., 2007; Selin et al., 2008).

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2.3. Emissions: the authors should make it clearer throughout that they are only looking at Hg(0) emissions in China (and not Hg(II) or Hg(P) emitted directly). It is obvious when comparing the numbers to the Pacyna reference, but readers not familiar with total Hg emissions in China may confuse this as it is not stated in the paper. It is not, as stated, that "All surface emissions are assumed to be in all the form of Hg(0)"; but that emissions in the inventories from Hg(II) and Hg(P) are omitted [correct?]

The uniform distribution of Hg(0) emissions from land and ocean is problematic, as land emissions are likely to have been affected by previous deposition and ocean emissions vary latitudinally. Though I agree with the authors that it plays little role in the present application, it would dramatically affect the agreement of model to measurements and thus may need to be improved for this purpose.

It would also be helpful in this section (perhaps in a table?) to detail the actual (Mg) emission from China under the different scenarios and include a few words about why they are different.

3.1. Results: Elemental Mercury It is impossible to see in Figure 2, given the color scale, whether the Hg(0) concentrations are remotely reasonable. The green color covers a range from 1.0-2.5 ng m⁻³. In addition, the authors do not give a global mean Hg(0) value or compare with any seasonal or annual mean data except for Athens, Ohio (Figure 3). It is also unclear in Fig 3 which is the data and what the circles and bar plots represent. Thus, as a reviewer I have no way of knowing whether the simulation is remotely reasonable. Key information which would be necessary in this regard are the following: a) the global budget of mercury in the model, including the lifetime of Hg(0) in the atmosphere; b) comparison with annual average Hg(0); c) whether there is an interhemispheric gradient and if so how large.

Page 19868 lines 12+: I am very confused as to why "gridding distributions" would cause differences in the Southern hemisphere. As is stated earlier, all of the simulations used the same inventories outside of China, so why should they be gridded

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differently other than due to human error? Also, why are there differences in the U.S.?

Page 19869 lines 1-15: It would be helpful to show the total concentrations which can be compared directly to the Jaffe data, to convince the reader that the model is reasonably simulating these concentrations, in addition to the China component.

same page....line 23: It is surprising to me that the r^2 is 0.8 without any Asian component. What does this suggest about the sources of Hg and CO in that case? Much more discussion is needed here and comparisons to Strode et al., who did a much more detailed analysis of the Okinawa data.

3.2.1. Results: Wet deposition P. 19870 line 6: "Wet deposition is highly seasonal..."; I assume that the authors mean in the U.S. There is no data elsewhere (e.g. the tropics) to support this statement.

The authors do no comparisons with total wet deposition measurements, for which there is an extensive data set in the U.S. Further, the authors note that while measurement data is low in winter in the U.S., calculated deposition is high during that time. The authors should discuss their parameterization of scavenging of Hg in snow in particular (see Mason et al., 2000). If the model does not correctly represent the processes of wet deposition over the United States, it is unclear whether the analysis of Asian sources is to be believed, as previous model studies have shown the strong role of scavenging from the free troposphere (see Strode et al., 2008; Selin and Jacob, 2008). A better discussion of transport and mixing in the free troposphere is needed here.

3.2.2 Dry Deposition: Here, and above, it would be helpful to be clearer in the language a) what the model predicts as the total Asian contribution, with a range between Streets and Pacyna, and b) what the uncertainty between Streets and Pacyna are. The way it is written with the differences between the two cases is confusing. I would suggest, here and elsewhere, focusing the language on what these cases are diagnosing rather than merely reporting their difference.

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4. Conclusions: The authors address aqueous chemistry for the first time here. It should be addressed and its implications more fully discussed above, in the mercury chemistry section.

Technical Corrections

The nomenclature of HGO for oxidized/reactive mercury is a bit confusing, as it is easy to confuse it with HgO (i.e. mercuric oxide). The more standard notation is Hg(II), and I suggest that this be used. I understand that in this case the authors refer to HGO as also including the particulate-bound fraction, but as this fraction is usually in the oxidized phase I think that the use of Hg(II) would still be warranted here.

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